

Polarization Dependence of SFG spectra of H/C(100) Surface

A. Wada^{a)}, H. Takaba^{a)}, T. Ando^{b)}, M. N. Gamo^{b)},
Y. Sato^{b)}, and C. Hirose^{b)}
(awada@csd.res.titech.ac.jp)

^{a)} Chemical Resources Laboratory*,
Tokyo Institute of Technology,
4259, Nagatsuta, Midori-ku, Yokohama, 226-8503, Japan.

^{b)} Core Research for Evolutional Science and Technology
(CREST) of Japan Science and Technology Corporation,
c/o National Institute for Research in Inorganic Materials
(NIRIM),
1-1 Namiki, Tsukuba, Ibaraki 305-0044, Japan

Chemical vapor deposition (CVD) is the most well-developed technique used to fabricate diamond films. In diamond film growth by CVD method, hydrogen atom is crucial for the enhancement of the growth rate by creating vacant sites through abstraction and for the improvement of the quality of the diamond film by suppressing graphite formation. Hydrogen-terminated diamond surfaces have attracted much attention because of the roles played by the hydrogen atoms in the CVD process.

For hydrogen-terminated diamond (100) surface, an energy loss peak was observed on HREEL spectrum at 2928 cm^{-1} which was assigned to the CH stretching band of monohydride (CH) group [1]. However, result of an *ab initio* self-consistent field molecular orbital calculation using a C_9H_{14} cluster predicted that the CH stretching vibration of monohydride group consists of two modes, one at 2920 cm^{-1} and one at 2903 cm^{-1} , because the diamond C(100)-2 \times 1 surface has dimer (HC-CH) rows and they should have two stretching vibration modes: in-phase stretching (symmetric stretching) and out-of-phase stretching (antisymmetric stretching) modes [2]. Recently, Takaba *et al.* observed the vibrational SFG peaks at 2924 cm^{-1} and 2899 cm^{-1} . From the peak frequencies and the input (visible and IR beams) polarization characteristics of the SFG peak intensities, the peaks were assigned to CH symmetric (2924 cm^{-1}) and CH antisymmetric (2899 cm^{-1}) stretching modes of HC-CH group [3].

In this paper, we report on the input (visible and IR beams) and output (SFG beam) polarization characteristics of the SFG spectral feature on a H/C(100)-2 \times 1 surface. On the SFG spectra, in addition to the vibrational resonance peaks at 2899 and 2924 cm^{-1} , the broad peaks were observed at around 2830 and 2930 cm^{-1} . We found that the vibrational resonance peaks showed the different polarization characteristics from the broad peaks.

High-pressure high-temperature (HP-HT) synthetic diamond C(100)-2 \times 1 surface prepared by M. N.-G., Y. S. and T. A. at NIRIM, was investigated. Hydrogenation was performed at 1073 K in hydrogen plasma generated by a 2.45 GHz microwave discharge. The discharge condition was similar to that of CVD diamond growth. After the plasma treatment, the sample was transferred to a UHV chamber (base pressure; 1×10^{-10} Torr) and cleaned by annealing for 20 min at 773 K under evacuation. The SFG measurements were carried out under the UHV condition at room temperature.

The fundamental (1064 nm) output pulses from a mode-locked Nd:YAG laser (35 ps, 10 Hz) were used for the generation of frequency-tunable IR pulses and the second harmonic (532 nm) pulses for the SFG measurements. The visible and IR pulses with the energies of 40 and 120 $\mu\text{J}/\text{pulse}$, respectively, were focused on the sample surface with the incident angles of 40 and 50°, respectively. The generated SFG pulses were passed through dielectric filters and a monochromator, and detected by a photomultiplier tube.

Figure 1 shows the SF polarization dependence of SFG spectra of H/C(100)-2 \times 1 surface under the input polarization combinations of (ps), where the first and second letters in parenthesis denote the polarizations of

visible and infrared beams, respectively. It is seen from the figure that the spectral feature depends on the polarization of SF beam. When the p-polarized SF beam was monitored, the broad peaks at 2830 and 2930 cm^{-1} were observed. On the other hand, the vibrational resonance peak at 2899 cm^{-1} was observed by detecting the s-polarized SFG signal. It was found from the SF polarization dependence on the other input polarization combinations, such as (pp), (sp), and (ss), the existence of the peaks at 2899 and 2924 cm^{-1} depends on the polarizations of IR and SF beam but the existence of the broad peaks depends on the polarizations of visible and SF beam. The results suggest that the origin of the broad peaks would not be the vibrational resonance.

- [1] T. Aizawa, T. Ando, M. Kamo, and Y. Sato, *Phys. Rev. B*, 48 (1993) 18348.
- [2] K. Ushizawa, M. N. -Gamo, Y. Kikuchi, I. Sakaguchi, Y. Sato, and T. Ando, *Phys. Rev. B*, 60 (1999) R5165.
- [3] H. Takaba, K. Kusafuka, M. N. Gamo, Y. Sato, T. Ando, J. Kubota, A. Wada, and C. Hirose, *Dia. and Relat. Mat.*, in press.

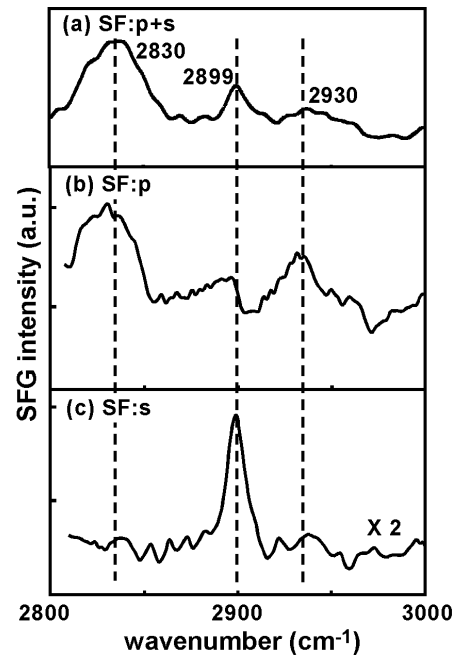


Fig. 1. SFG spectra of H/C(100) surface under the polarization condition of visible-p and IR-s. Polarization of SF light is (a) p and s, (b) p, and (c) s.